Double-Pulse Deexcitations in a One-Dimensional Strongly Correlated System

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We investigate the ultrafast optical response of the one-dimensional half-filled extended Hubbard model exposed to two successive laser pulses. By using the time-dependent Lanczos method, we find that following the first pulse, the excitation and deexcitation process between the ground state and excitonic states can be precisely controlled by the relative temporal displacement of the pulses. The underlying physics can be understood in terms of a modified Rabi model. Our simulations clearly demonstrate the controllability of ultrafast transition between excited and deexcited phases in strongly correlated electron systems.

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Coherent manipulations of quantum states are essential prerequisites for many advanced technological applications. One of the fundamental phenomena in the field of nonlinear light-matter interactions are Rabi oscillations describing the time evolution of a two-level quantum system exposed to an external field. The Rabi oscillation and its generalizations, together with related concept of quantum interference, play central roles in the fields of coherent control and manipulations. In decades, with the development of ultrafast optical techniques, quantum interference experiments have advanced beyond atomic/molecular systems into more complex solid state systems, such as semiconductor heterostructures and quantum dots [1–5].

Recently, there has been a steady increase in research and development of materials with correlated electrons, due to their multiple novel features that will potentially provide key ingredients for future technological applications and innovations. For example, resulting from the competing orders inherent in correlated systems [6], it is plausible to obtain significant response, and induce fast transitions from one phase to another with relatively weak external pulse [7]. In order to achieve efficient high performance manipulations, it is imperative to examine the ultrafast coherent dynamics of these systems when subjected to external stimuli. In this Letter we show that under specific conditions Rabi-like oscillations may be observed in a prototype correlated system close to the charge density wave (CDW) transition.

The one-dimensional (1D) Mott insulators, including Sr_2CuO_3 and halogen-bridge Ni compounds [8] as their well-known representatives, have been considered as promising candidates for future optoelectronic materials. Swift recoveries (within picoseconds) to the insulating state in the photoinduced insulator-to-metal transitions have been noticed [9, 10], which can be three orders of magnitude faster than in semiconductors [11]. Gigantic third-order optical nonlinearity has also been reported [12]. A good theoretical starting point to study 1D Mott insulators is the extended Hubbard model [13]. In the later, besides the on-site repulsion U, nearest neighbor electron-electron interactions, denoted as V, are also included. It is well known that when V exceeds a critical value, bound states of charge carriers, i.e., doublons (double-occupied sites) and holes (empty sites), are stabilized [14, 15]. In some region of the interaction parameter space, the contribution to the optical absorption spectrum from these bound states, known as excitons, can give rise to an isolated δ-like peak with energy below the Mott gap. The influence of the low-lying excitonic excitations on optical properties has been examined theoretically [16, 17] and experimentally [18]. Nevertheless, the role of the exciton in the ultrafast optical dynamics has not been fully addressed.

Using the time-dependent Lanczos method, we investigate the extended Hubbard model at half filling. We are particularly interested in its optical response to an external field composed of two consecutive laser pulses, which resemble the pump-probe setup in spectroscopy experiments. Following the time evolution of the system in the ultrafast regime, we show that by carefully tuning the interval between the two pulses, the system can be excited by the first pulse and then deexcited by the second one. The conditions for observing such on-and-off phenomenon and the underlying physics are analyzed. More specifically, close to the CDW phase transition where long-lived excitons represent optically active excitations, the many-body system can be mapped onto an effective two-level system, and accordingly, its dynamical response can be understood in terms of a simple modified Rabi model. Alternatively, far from the CDW phase transition, e.g., $V = 0$, such on-and-off signal is diminished by the continuity of its optical spectrum and the fast decay of charge carriers into the continuum. This

controllable, ultrafast switching realized in this model, though conceptually simple, can be easily generalized to other correlated systems as well, providing that certain type of excitations can be selectively excited by carefully tuned optical pumps.

The half-filled extended 1D Hubbard model is written as

$$H = -t_h \sum_{i,\sigma} \left(c^\dagger_{i,\sigma} c_{i+1,\sigma} + \text{H.c.} \right) + U \sum_i \left(n_{i,\uparrow} - \frac{1}{2} \right) \times \left(n_{i,\downarrow} - \frac{1}{2} \right) + V \sum_i \left(n_i - 1 \right) \left(n_{i+1} - 1 \right), \quad (1)$$

where $c^\dagger_{i,\sigma}$ ($c_{i,\sigma}$) is the creation (annihilation) operator for an electron with spin σ at site i, $n_i = n_{i,\uparrow} + n_{i,\downarrow}$, t_h is the hopping constant, U and V are on-site and nearest-neighbor repulsion strength, respectively. As V increases, the charge carriers, i.e., doublons and holes, become energetically more favorable, since V mediates an effective attraction between holes and doublons. It has been shown that in large-U limit, when V exceeds a critical value $\sim 2t_h$, excitonic bound states are formed in the photoexcitation processes [14, 15], while the signature of these bound states emerges as a peak-like structure at the lower edge of the optical spectrum, separated from the continuum. The strength of the peak is enhanced with the increase of V [16, 17]. At large U, a first-order phase transition from the spin-density-wave (SDW) to the CDW phase takes place when $V_c \approx U/2$ [19, 20].

The external laser pulse is introduced via the Peierls substitution in the hopping terms of the Hamiltonian (1):

$$c^\dagger_{i,\sigma} c_{i+1,\sigma} \to e^{iA(t)} c^\dagger_{i,\sigma} c_{i+1,\sigma}, \qquad H \to H(t). \quad (2)$$

The time-dependent vector potential $A(t)$ is written in the temporal gauge [21–23]:

$$A(t) = A_0 e^{-(t-t_1)^2/2t_d^2} \cos\left[\omega_{\text{pump}} (t - t_1)\right] + A_0 e^{-(t-t_2)^2/2t_d^2} \cos\left[\omega_{\text{pump}} (t - t_2)\right]. \quad (3)$$

Here, for simplicity, two pulses with identical shape are assumed. Their temporal separation is given by $\Delta t := t_2 - t_1$, the distance between the two peaks. A_0 controls the laser intensity, t_d determines the widths of pulses, and ω_{pump} is the pumping frequency.

In order to investigate the system's evolution under the influence of the pulses, we employ the time-dependent Lanczos method, which is originally described in Ref. [24], and has been developed into one of the standard numerical methods in the study of nonequilibrium dynamics of correlated systems [25]. The basic idea is that starting from the Schrödinger equation $i\partial\psi(t)/\partial t = H(t)\psi(t)$ (we have set $\hbar \equiv 1$), the time evolution of the wave function $|\psi(t)\rangle$ is approximated by a step-vise change of time t into small increments δt. At each step, the Lanczos basis with dimension M is generated, resulting in the time evolution

$$|\psi(t+\delta t)\rangle \simeq e^{-iH(t)\delta t}|\psi(t)\rangle \simeq \sum_{l=1}^{M} e^{-i\epsilon_l \delta t}|\phi_l\rangle\langle\phi_l|\psi(t)\rangle, \quad (4)$$

where ϵ_l and $|\phi_l\rangle$, respectively, are eigenvalues and eigenvectors of the tridiagonal matrix using M Lanczos iterations.

In the following discussions, we set t_h and $1/t_h$ as energy and time units, and we fix $U = 10\,t_h$. Periodic boundary conditions are imposed. The ground state in all cases represents the initial state, i.e., $|\psi(t=0)\rangle = |\text{GS}\rangle$. The time increment $\delta t = 0.01$, and the Lanczos dimension cutoff is set to $M = 30$.

We carry out numerical simulations for the Hamiltonian (2) at half filling. Here, we focus on the time dependence of the energy in the resonance case, i.e., the pumping frequency ω_{pump} matches the positions of the absorption peaks in the optical spectra. The time-dependent energy is defined as

$$E(t) = \langle\psi(t)|H(t)|\psi(t)\rangle - E_{\text{GS}}. \quad (5)$$

Our main results are shown in Fig. 1 for 14 lattice sites at $V = 4.5$, where the ground state is in the SDW phase. The exciton state is an excited state with the energy $\omega_{\text{R}} = 4.0$ above the ground state for this particular system size, and we put $\omega_{\text{pump}} = \omega_{\text{R}}$. We deliberately chose a small value of $t_d = 0.65$, while the reason will be given later. The shape of $A(t)$ is shown in Figs. 1(a) and

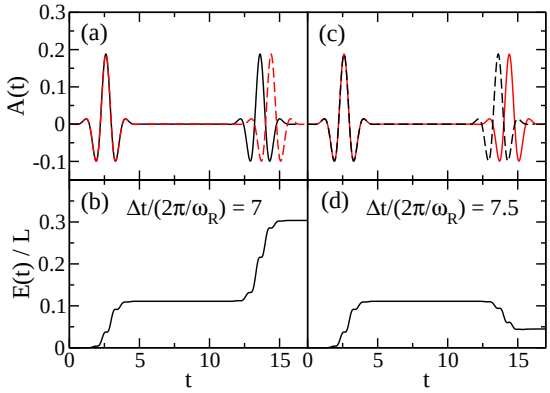

FIG. 1. (Color online) Time dependence of the vector potential $A(t)$ ((a) and (c)) and the energy (measured from the ground state) per site $E(t)/L$ (L is the lattice size) under the action of two consecutive pulses ((b) and (d)), for 14-site lattice with $U = 10$, $V = 4.5$. Parameters for the laser pumps: $A_0 = 0.19$, $\omega_{\text{pump}} = \omega_{\text{R}} = 4.0$, $t_d = 0.65$. The first peak of the pulses is fixed at $t_1 = 4\,t_d$; the second peak is determined by $t_2 = t_1 + \Delta t$. $\Delta t = 7$ ((a) and (b)) and 7.5 ((c) and (d)), respectively, in the unit of $2\pi/\omega_{\text{R}}$. In the upper part, the solid lines represent the shapes of the corresponding vector potential $A(t)$, while the other case is shown as dash line for comparison. The results of $E(t)$ are shown in the lower part.

(c). Suppose that $t_h = 1$ eV, then in the real time unit, $\hbar/t_h = 0.66$ fs, $t_d = 0.43$ fs. The intensity of the laser pulse $A_0 = 0.19$ is used so that the number of photons (per site) during the pulse, $N_{\rm pt} \propto A_0^2 \omega_{\rm pump} t_d$, is around 0.5 [23], which means that we confine our calculations in the weak excitation regime [5]. The maximum of the electric field can reach around 7.6×10^9 V/m if the lattice constant is 1 Å.

The main message in Fig. 1 is that by tuning Δt, the final energy $E_{\rm fin}$ after the second pulse can be either enhanced (Fig. 1(b)) or reduced (Fig. 1(d)), compared with the intermediate stage energy (the energy between the two pulses, which can be estimated from the plateau of $E(t)$ in Fig. 1). It turns out that the beat is controlled by the energy difference between the ground state and the excited states, i.e., $\omega_{\rm R}$. As shown in Fig. 1, we notice that if $\Delta t = N \times \frac{2\pi}{\omega_{\rm R}}$, where $N \in$ Integer, the highest enhancement is found; on the other hand, the strongest suppression appears when $\Delta t = \left(N + \frac{1}{2}\right) \times \frac{2\pi}{\omega_{\rm R}}$. It is noted that by tuning parameters, we are able to obtain almost perfect energy suppression after the second pulse. For example, for $t_d = 5$, $A_0 = 0.01$, the corresponding $N_{\rm pt} \sim 0.01$, $E_{\rm fin}$ turns out to be 0.003.

The underlying physical picture can be captured by the Rabi model [26, 27]. The latter describes a two-level quantum system coupled with a single-mode external field, and can be written as

$$H_{\rm R}(t) = \epsilon \sigma_z + g(t) \sigma_x, \tag{6}$$

where σ_a are Pauli matrices, 2ϵ is the level spacing; the off-diagonal term $g(t)$, which describes the coupling of the two-level system to an external field, typically has the form $g(t) = 2g \cos \omega t$. When $\omega \approx 2\epsilon$, the rotating-wave approximation can be applied [28]. The system's oscillation between the two levels, known as Rabi oscillation, is fully characterized by two quantities, δ and Ω_R, known as detuning and Rabi frequencies, respectively:

$$\delta = \epsilon - \omega/2, \quad \Omega_R = \left[(\epsilon - \omega/2)^2 + g^2\right]^{1/2}. \tag{7}$$

At the tuning point, i.e., $\omega = 2\epsilon$, only Ω_R remains, and is solely determined by the coupling strength g.

Keeping this picture in mind, we extend this model to a case when $g(t)$ in Eq. (6) is substituted by $A(t)$ (Eq. (3)). Figure 2 shows the energy expectation of the modified Rabi model at the tuning frequency $\omega_{\rm pump} = 2\epsilon$. A typical quantum interference phenomenon is observed. We can see that after a coherent exciton polarization is created by the first pulse, the system energy $E(t)$ is either raised (Fig. 2(a)) or reduced (Fig. 2(b)) by the second pulse, depending on the relative phase of the two pulses [4].

The plain similarity between Figs. 1 and 2 suggests that the two-level model can provide qualitative understanding of the optical response of the extended Hubbard

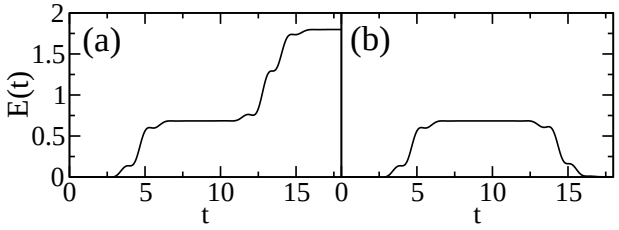

FIG. 2. The time-dependent energy expectation $E(t)$ (measured from the lower level) for the Rabi model coupled with two successive pulses. Here, we take $\epsilon = 1$. Parameters for $A(t)$: $A_0 = 0.5$, $\omega_{\rm pump} = 2\epsilon = 2$, $t_d = 1$, $t_1 = 4t_d$. In (a), $\Delta t = 3 \times \pi/\epsilon$; (b) $\Delta t = 3.5 \times \pi/\epsilon$.

model. The analogy is justified by the existence of excitonic peak in the optical absorption spectrum [17] for the given value of $V = 4.5$, which is sharp and isolated from the continuum. For this reason the decay time of the excitonic state is much slower than Δt which enables nearly complete deexcitation after the second external pulse. It should be noted that the study on coherent dynamics between several inherent energy levels under the influence of the external stimuli is a widespread practice as mentioned in the introduction. Yet to our knowledge, quantitative investigations concerning the possibility of this kind of setup on correlated systems are few and far between.

Naturally, we may wonder what will happen in the case of $V = 0$, where instead of well-defined excitonic peak, a continuum absorption is expected in the thermodynamic limit, reflecting the response from unbound charge carriers. In order to answer this question, we perform the calculation on various lattice sizes, $L = 10$, 12 and 14. Due to the finite-size effects, a series of separate peaks with comparable heights, representing a continuum in the thermodynamic limit, appear in the optical absorption spectrum. The number of large peaks scales linearly with the system size L, which can be interpreted as the finite-size precursor of the continuum. Based on this observation, our strategy is the following: we set the center of pumping frequency $\omega_{\rm pump}$ to match the first major peak in the optical spectrum, denoted as ω_1, while t_d is determined by the difference between the first and second resonance frequencies, i.e., $t_d = 1/(\omega_2 - \omega_1)$. This is the same approach as we have already used for the calculations of Fig. 1. In this way, the multi-frequency absorption, which is expected in the thermodynamic limit when $V = 0$, can be approximately simulated. For various lattice sizes, we use identical $N_{\rm pt} \sim 0.5$. The laser intensity A_0 can then be determined accordingly.

Figure 3 shows the final energy per site $E_{\rm fin}/L$ as a function of the normalized temporal interval $\tilde{\Delta}t$, for various lattice sizes $L = 10$, 12 and 14. In Fig. 3(a) we present results for $V = 0$, and in (b) for $V = 4.5$, with $\tilde{\Delta}t := \Delta t/(2\pi/\omega_1)$ and $\Delta t/(2\pi/\omega_{\rm R})$, respectively. The

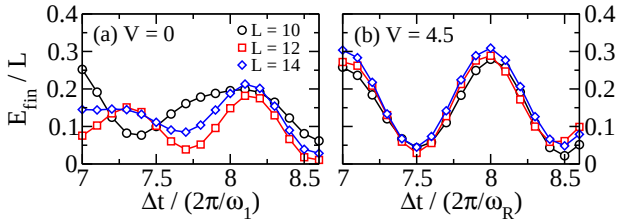

FIG. 3. (Color online) The final energy per site after the action of the second pulse, $E_{\rm fin}/L$, as a function of Δt for various lattice sizes $L = 10$, 12 and 14. (a) for $V = 0$; (b) $V = 4.5$. Δt is normalized by $2\pi/\omega_1$ and $2\pi/\omega_{\rm R}$, respectively. In all cases, $N_{\rm pt}$ is fixed to 0.5. The statements on how to specify other parameters, i.e., $\omega_{\rm pump}$, t_d, and A_0, can be found in the main text.

results for $\tilde{\Delta}t$ ranged from 7 to 8.6 are presented. In clear contrast to the almost perfect periodicity at $V = 4.5$ (Fig. 3(b), where peaks (valleys) of $E_{\rm fin}$ are found independently of L at the integer (half-integer) values of $\tilde{\Delta}t$), Fig. 3(a) for $V = 0$ shows a different picture. Results for different $L = 10$ through 14 are consistent with the admixture of higher harmonics and display large finite-size effects. At $\tilde{\Delta}t = 7$, large differences for different L are found. After $\tilde{\Delta}t = 8$, the curves show more consistent results. However, with increase of $\tilde{\Delta}t$, results for $E_{\rm fin}$ start showing larger discrepancies again, e.g., around $\tilde{\Delta}t = 11$ (not shown).

We suggest that there could be two reasons responsible for the irregularities observed in Fig. 3(a). One is the mixing of absorptions at different frequencies. Here, according to our setting, at least two resonance frequencies are in the reach of the pumping pulse. We can expect that with the increase of the system size separated peaks in the optical spectrum gradually form a continuum. Therefore, for a given t_d, the distribution of optically-allowed states become continuous within the reach of pumping frequencies, which in turn gradually diminishes the excited-deexcited oscillations that appear in small-size calculations. Another reason might be the decay of charge carrier pairs, due to the lack of the protection of bound states. Let us suppose that a doublon-hole pair is created, separately by a single lattice site. Subsequently, hopping of the doublon (or the hole) to the neighboring sites leads to an incoherent decay of the excitation into the doublon-hole continuum [29]. Compared with Fig. 3(b), we speculate that it can be the main underlying mechanism for the disparity between adjacent cycles found in Fig. 3(a).

In summary, we propose an optical interferometric experiment to detect Rabi-like oscillations in a strongly correlated system, i.e., the extended Hubbard model. Depending on the relative phase, it is possible to observe the system being excited and then deexcited (or enhanced further) by two consecutive laser pulses. A comparative study of the Hubbard model without nearest neighbor interactions highlights the important role played by the excitonic bound states in this ultrafast dynamics. The key finding of this letter is that when a precisely selected electric pulse in a correlated system triggers the excitation of a many-body state that has no efficient decay path, a quantum interference measurement may be realized within the experimentally accessible time scale. Suggested experiments may lay course for further research of coherent control and manipulations on many-body systems.

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